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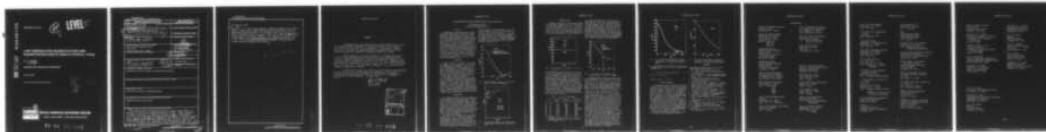
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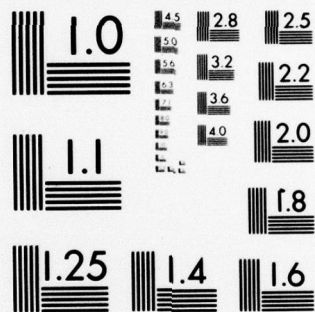
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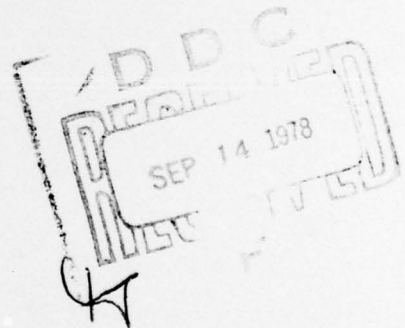
LOW TEMPERATURE MAGNETIZATION AND MAGNETOSTRICTION OF SINGLE CRYSTAL $TmFe_2$

BY R. ABBUNDI
A. E. CLARK

RESEARCH AND TECHNOLOGY DEPARTMENT

17 JULY 1978

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erg/cm³ at 0 K. The saturated magnetic moment was found to be 74 emu/g, which is substantially larger than previous polycrystalline results. Magnetostriction measurements are consistent with $|\lambda_{111}| \gg |\lambda_{100}|$, and yield a saturation value of $\lambda_{111}(0) = -3520 \times 10^{-6}$, which is appreciably larger than that previously reported. The rapid fall of the T_m sublattice moment with temperature results in a rather low value of $\lambda_{111} = -210 \times 10^{-6}$ at 300K. The temperature dependence of λ_{111} closely follows single-ion magnetoelastic theory.

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SUMMARY

The magnetization and magnetostriction study reported here is part of a research program undertaken to determine the nature of the magnetostriction in the rare earth (R)-Fe₂ compounds. In this paper is detailed the correlation between the magnetostriction and magnetization for TmFe₂. Studies were made as a function of temperature from 4°K to 300°K.

TmFe₂, because of its highly anisotropic 4f charge distribution, displays a huge magnetocrystalline anisotropy (K_1) and magnetostriction (λ_{111}) at cryogenic temperatures in spite of the high cubic symmetry of the Cl5 lattice and the tetrahedral symmetry of the Tm³⁺ site. Magnetostriction measurements are consistent with $|\lambda_{111}| \gg |\lambda_{100}|$, and yield a saturation value of $\lambda_{111} = -3520 \times 10^{-6}$. The temperature dependence of λ_{111} closely follows single-ion magnetoelastic theory.

The study was carried out in the Solid State Branch of the Radiation Division as part of the research program on magnetostrictive material. The research was sponsored by the Office of Naval Research (PO 4-0081, NR 039-110) and the NSWC Independent Research Funds.

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LOW TEMPERATURE MAGNETIZATION AND MAGNETOSTRICTION OF SINGLE CRYSTAL TmFe_2 *

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Naval Surface Weapons Center
White Oak, Silver Spring, MD 20910

ABSTRACT

The magnetization and magnetostriction of single crystal TmFe_2 were measured as a function of temperature from 4K to 300K. TmFe_2 , because of its highly anisotropic 4f charge distribution, displays a huge magnetocrystalline anisotropy (K_1) and magnetostriction (λ_{111}) at cryogenic temperatures in spite of the high cubic symmetry of the Cl_5 lattice and the tetrahedral symmetry of the Tm^{3+} site. The anisotropy spans the range from -5.3×10^5 erg/cm³ at room temperature to the extrapolated value of -5×10^8 erg/cm³ at 0 K. The saturated magnetic moment was found to be 74 emu/g, which is substantially larger than previous polycrystalline results. Magnetostriction measurements are consistent with $|\lambda_{111}| \gg |\lambda_{100}|$, and yield a saturation value of $\lambda_{111}(0) = -3520 \times 10^{-6}$, which is appreciably larger than that previously reported. The rapid fall of the Tm^{3+} sublattice moment with temperature results in a rather low value of $\lambda_{111} = -210 \times 10^{-6}$ at 300K. The temperature dependence of λ_{111} closely follows single-ion magnetoelastic theory.

INTRODUCTION

Despite the overall cubic symmetry and the high local symmetry at the rare earth site, the rare earth- Fe_2 compounds possess huge magnetostrictions and magnetic anisotropies, which extend, in many cases, as high as room temperature. The compounds TbFe_2 and SmFe_2 yield magnetostrictions: $|\lambda(300\text{K})| > 2000$ ppm. [1] TbFe_2 and DyFe_2 possess anisotropies: $|K_1(300\text{K})| > 10^7$ erg/cm³. [1,2] These compounds have relatively saturated rare earth magnetizations at room temperature, thus only moderate increases in magnetostriction and anisotropy occur at reduced temperatures.

TmFe_2 however, possesses a rather small magnetostriction and magnetic anisotropy at room temperature. Here we show that this is directly correlated with the rather low value of the Tm^{3+} sublattice moment at room temperature. In this paper, we report magnetization measurements on single crystal TmFe_2 . From the anisotropy in the moment we determine the magnetocrystalline anisotropy, K_1 . Finally we measure λ_{111} for TmFe_2 from 6-300 K. Temperature dependences are compared to those calculated from single-ion theory. Intrinsic values are compared to those of other RFe_2 compounds using Stevens' equivalent operator coefficients.

MAGNETIZATION

The ferrimagnetic nature of TmFe_2 was identified by Wallace and Skrabek [3] and by Burzo [4] with magnetization measurements on polycrystalline samples. A compensation temperature was observed in TmFe_2 at approximately 240 K, indicating a rapid drop of Tm^{3+} sublattice moment with increasing temperature. In addition to this rapid decrease, the intrinsic ($T=0$) moment itself was reported to be very low, too low to be consistent with reasonable assumptions of rare earth and iron sublattice moments. Because of the potentially high anisotropy of TmFe_2 inferred from measurements on other RFe_2 compounds, [2] such a low value could result from non-isotropic polycrystalline samples.

The single crystals of TmFe_2 used in this experiment were grown by O. D. McMasters by horizontal zone techniques. [5] In Fig. 1, we show the spontaneous magnetic moment of single crystal TmFe_2 along its crystallographic [111] easy axis. While the room temperature moment is similar to that observed earlier in polycrystals by Wallace [3] and Burzo, [4] the low-temperature moment ($\sigma_g(0)$) is substantially larger. Fig. 2 illustrates the magnetization curves along the three

principal directions at room temperature. A huge magnetic anisotropy is present at low temperatures. Taking the saturation moment from Fig. 1 to be 74 emu/g ($3.72\mu_B$) and a gJ value of $7\mu_B$ for Tm^{3+} , we calculate a Fe moment of $1.64\mu_B$, in good agreement with that of other RFe_2 compounds. [4,6,7] A rare earth moment, μ_R , less than $7\mu_B$, would result in a correspondingly smaller iron moment, μ_{Fe} . In the single crystal, the compensation point is shifted only slightly from the earlier measurements on polycrystals.

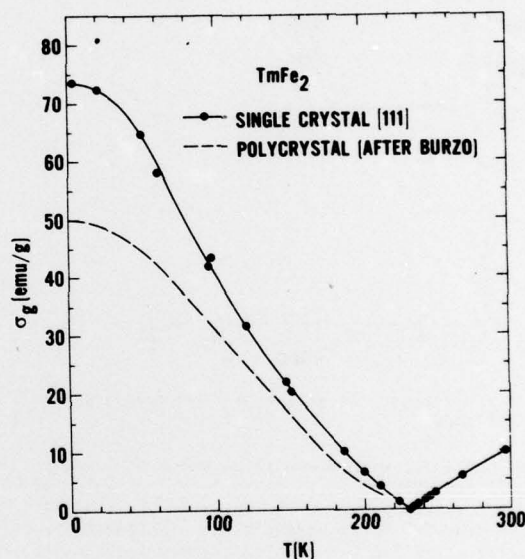


Fig. 1 Spontaneous magnetic moment of single crystal TmFe_2 as a function of temperature.

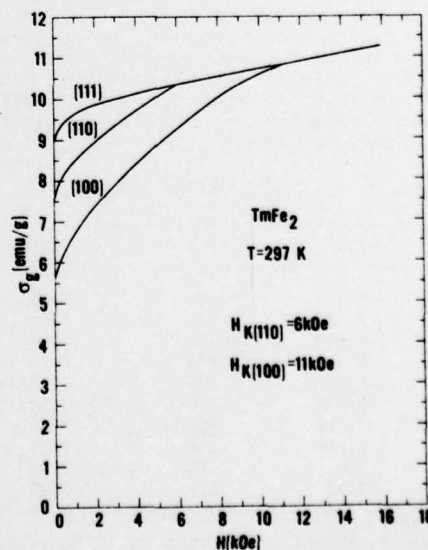


Fig. 2 Room temperature magnetization along principal crystallographic directions of single crystal TmFe_2 .

MAGNETOSTRICTION

Because of the rapid fall of the sublattice moment with temperature, we expect a very low value of magnetostriction at room temperature for TmFe_2 . Indeed this is the case for polycrystalline TmFe_2 : $\lambda_s = -123 \times 10^{-6}$. [1] The single crystal value, λ_{111} , is not much higher (see Fig. 3). Since $\lambda_{111} \gg \lambda_{100}$ for the $\text{Tb}_x\text{Dy}_{1-x}\text{Fe}_2$ ternary alloys, [8] it is plausible that here $\lambda_{111} \gg \lambda_{100}$; hence $\lambda_s \approx .6 \lambda_{111}$ for TmFe_2 . From measurements at 6 K we find the saturation value of $\lambda_{111}(0) = -3520 \times 10^{-6}$.

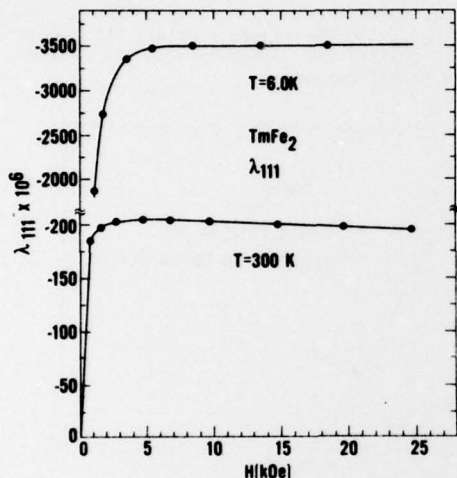


Fig. 3 λ_{111} of single crystal TmFe_2 at $T = 6.0$ K and $T = 300$ K.

This does not agree with the earlier value of -2600×10^{-6} reported by Barbara. [9] In Table I, our value of $\lambda_{111}(0)$ is compared to the value for TbFe_2 , estimated previously from 100 K measurements. [10] Following Stevens; [11] $\lambda_{111}(0) = C \alpha J(J+1/2) \langle r_F^2 \rangle$, where J is the angular momentum quantum number, α is the "Stevens' factor" and $\langle r_F^2 \rangle$ is the average radius squared of the 4f electron cloud. [12] The agreement between TmFe_2 and TbFe_2 is excellent. Taking $C = -0.017 \pm 10\% a_0^2$ the intrinsic magnetostrictions of the R^{3+} ions in the RFe_2 lattice are calculated. Note that the largest "+" magnetostrictions are predicted for CeFe_2 , PrFe_2 , TbFe_2 and DyFe_2 , and the largest "-" magnetostrictions for SmFe_2 , TmFe_2 and YbFe_2 . Since Ce is essentially always quadrivalent; Yb, divalent; and PrFe_2 does not crystallize into Laves phase structure, the remaining compounds are TbFe_2 , DyFe_2 , SmFe_2 and TmFe_2 .

Table I. $\lambda_{111}(0)$ of R^{3+}Fe_2 Compounds

R	J	$\alpha \times 10^2$	$\langle r_F^2 \rangle (a_0^2)$	$\lambda_{111}(0) \times 10^6$
Ce	5/2	-5.72	1.20	5660
Pr	4	-2.10	1.086	5280
Nd	9/2	-0.643	1.001	1920
Sm	5/2	4.13	.883	-3010
Tb	6	-1.01	.756	4160(4400 ^b)
Dy	15/2	-0.635	.726	4000
Ho	8	-0.222	.696	1540
Er	15/2	0.254	.666	-1470
Tm	6	1.01	.639	-3520 ^a
Yb	7/2	3.18	.613	-3370

a. This paper.

b. A. Clark, R. Abbundi, H. Savage and O. McMasters, Physica 86-88B, 73 (1977).

A small forced magnetostriction is observed at all temperatures above 15 K. Fig. 4 illustrates this behavior near T_{comp} . Below 234 K, the magnetostriction increases with magnetic field as the rare earth sublattice becomes further aligned. Above T_{comp} , where the rare earth sublattice points antiparallel to the field, the magnetostriction decreases with increasing field. Near magnetization compensation, where $\sigma_R = \sigma_{\text{Fe}}$; it is difficult to rotate the moments in the sample from one crystallite direction to another. Thus the full magnetostriction is not sensed, resulting in a dip in observed values. Precisely at T_{comp} , the field is perpendicular to the moments, resulting in an effective magnetostriction (as referred to field axes) of opposite sign.

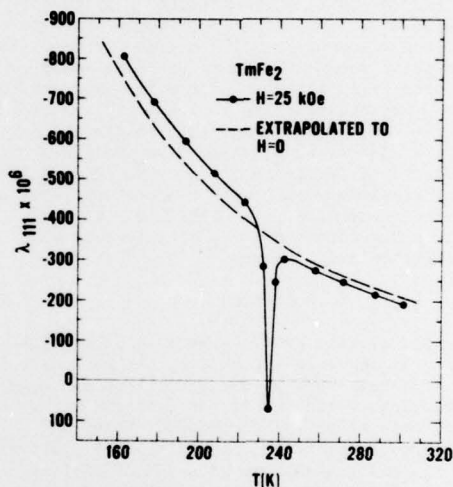


Fig. 4 λ_{111} of single crystal TmFe_2 near T_{comp} extrapolated to $H = 0$ and for $H = 25$ kOe.

It has been observed previously [10] that the temperature dependence of λ_{111} for TbFe_2 can be fit by the single-ion expression: $\lambda_{111} \propto \langle r_F^2 \rangle [\chi^{-1}(m_R)]$ of Callen and Callen [13] for $\ell=2$. Here χ^{-1} is a reduced Bessel function; χ^{-1} , the inverse Langevin function; and m_R , the reduced rare earth sublattice magnetization ($M_R(T)/M_R(0)$). In TbFe_2 the sublattice magnetization [9,14] drops by only $\sim 13\%$ and the temperature dependence follows closely the low temperature expansion: $m_R \approx m_R^3$. [10] In TmFe_2 , a good test of the single-ion expression over a wide temperature range can be obtained since, at 300 K, $m_R = .3$. To calculate the theoretical magnetostriction, it is necessary to know the normalized sublattice moment, m_R , as a function of temperature. This can be determined directly from neutron diffraction data, or from the total magnetization i.e. Fig. 1, subtracting off the contribution of the Fe sublattice. The currently known neutron diffraction data is not sufficiently accurate to determine $m_R(T)$ with the required precision. (The neutron diffraction data of both Rhyne [14] and Barbara [9] do not yield a compensation point in the correct temperature region.) Hence, here we shall determine $m_R(T)$ from the total magnetization curve (Fig. 1) minus $M_{\text{Fe}}(T)$, where $M_{\text{Fe}}(0)$ is estimated from the rare earth moment of gJ . In Fig. 5, we show the temperature dependence of λ_{111} from 0 K to 300 K. We assume a small temperature dependence for $M_{\text{Fe}}(T)$ consistent with a variety of sources. [14, 15,16] The dashed line is the calculated curve with $\mu_{\text{Fe}} = 1.64 \mu_B$ and a 15% reduction in Fe moment from 0 K to 300 K; the dash-dot line is a similar curve for a 20% reduction. A small ($\sim 3\%$) reduction of the rare earth moment from gJ , results in less than a few percent change in the calculated magnetostriction over the entire temperature range. The agreement between experiment and calculated curves is excellent.

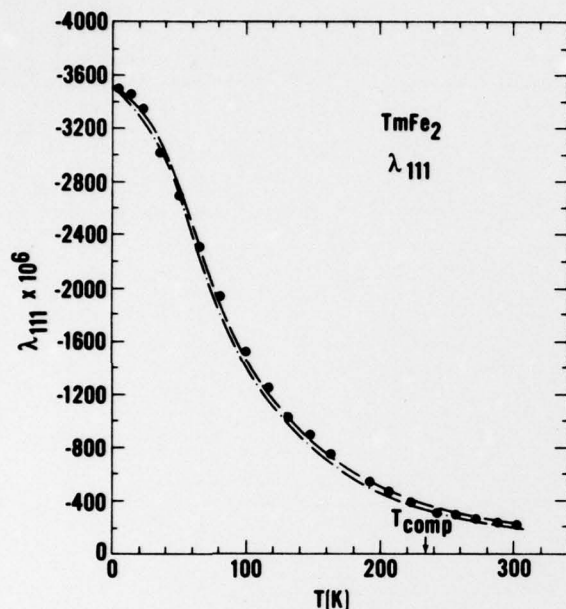


Fig. 5 λ_{111} as a function of temperature of single crystal TmFe_2 . The dashed curves represent the magnetostriction calculated from single-ion theory.

From the foregoing observations, we conclude that the magnetostriction, λ_{111} , of the RFe_2 compounds can be determined from:

$$\lambda_{111} = C \propto J(J-1/2) \langle r_F^2 \rangle \hat{I}_{5/2} [\mathcal{L}^{-1}(m_R)], \quad (1)$$

with $C = -0.017 \pm 10\% a_0^{-2}$.

$$\text{Since } \lambda_{111} \gg \lambda_{100}, \lambda_s \approx 3 \lambda_{111}/5. \quad (2)$$

MAGNETIC ANISOTROPY

The magnetization curves at room temperature (Fig. 2) reveal anisotropy fields H_{100} and H_{110} of 11 kOe and 6 kOe respectively. From these values we calculate room temperature anisotropy constants: $K_1 = -5.3 \times 10^8 \text{ erg/cm}^3$ and $|K_2| < 10^8 \text{ erg/cm}^3$. Similar curves are found for $T=200, 150$, and 97 K . Below 97 K , it was impossible to determine H_{100} and H_{110} with the available magnetic fields. In Fig. 6, $K_1 (\equiv MH_{100}/2)$ is plotted vs temperature and compared to the calculated temperature dependence arising from the single-ion model: $[13] \hat{I}_{\ell+1/2} [\mathcal{L}^{-1}(m_R)]$ where $\ell = 4$. The theoretical temperature dependence is in good agreement with the data, yielding an extrapolated value of $-K_1(0) \approx 5 \times 10^8 \text{ erg/cm}^3$.

According to the Stevens' equivalent operator coefficients, [11] the value of $K_1(0)$ varies according to $C B_J(J-1/2)(2J^2-5J+3)\langle r_F^2 \rangle$. This function does not depend strongly on rare earth ion. In agreement with this relationship, our extrapolated value of $-5 \times 10^8 \text{ erg/cm}^3$ for TmFe_2 is close to that of $-5 \times 10^8 \text{ erg/cm}^3$ for ErFe_2 [17] and -4.4×10^8 estimated for TbFe_2 . [2]

We gratefully acknowledge the help of W. Gillmor for the magnetic moment measurements of TmFe_2 .

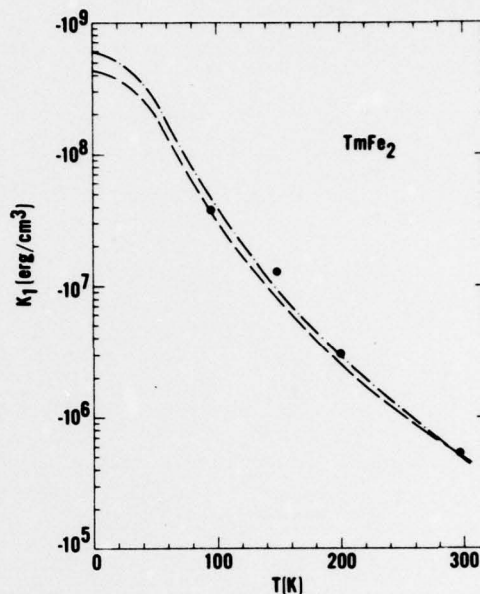


Fig. 6 K_1 of single crystal TmFe_2 as a function of temperature. The dashed curves represent the anisotropy calculated from single-ion theory.

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